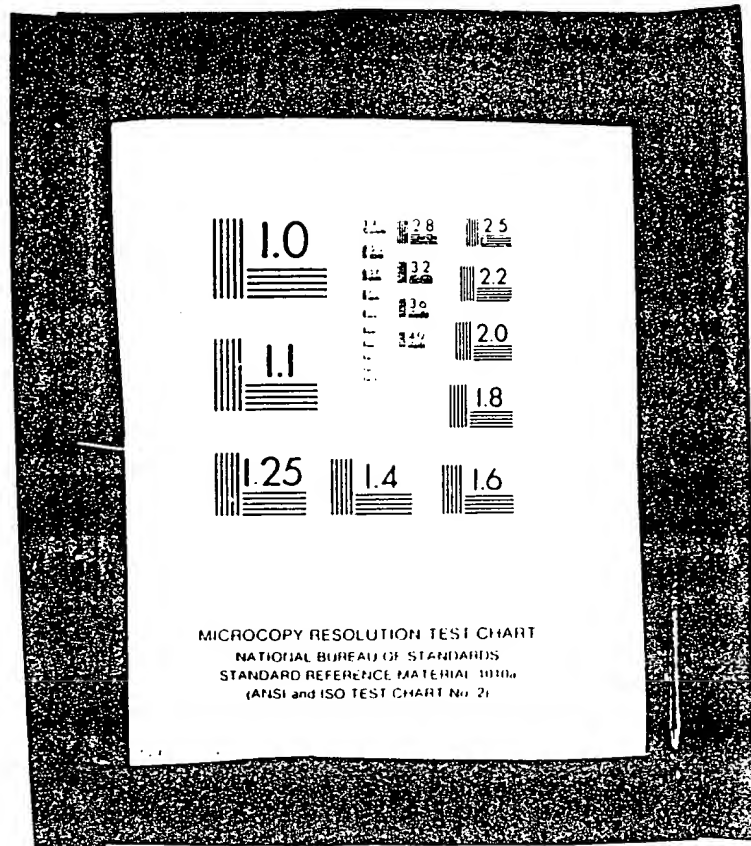
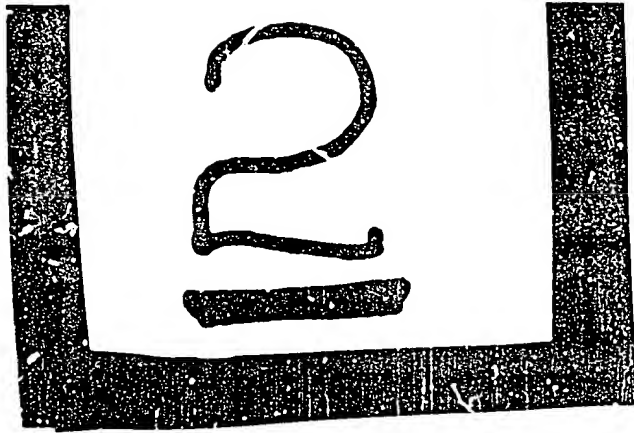




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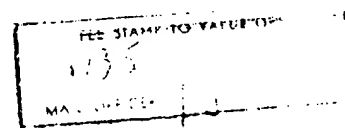


(CONVENTION. By one or more

COM



## CONVENTION APPLICATION FOR A PATENT



(1) Here  
insert (in  
full) Name  
of Name of  
Applicant or  
Applicants,  
followed by  
Address (es).

kx (1) HOECHST AKTIENGESELLSCHAFT

We  
of 45 Bruningstrasse, D6230 Frankfurt/Main 80,  
Federal Republic of Germany

(2) Here  
insert Title  
of Invention.

hereby apply for the grant of a Patent for an invention entitled (2)

POLYPROPYLENE MOLDING MATERIAL

(3) Here insert  
number(s)  
of basic  
application(s)

which is described in the accompanying complete specification. This application is a  
Convention application and is based on the application numbered (3)

P37 04 207.6

(4) Here insert  
Name of basic  
Country or  
Countries, and  
basic date or  
dates

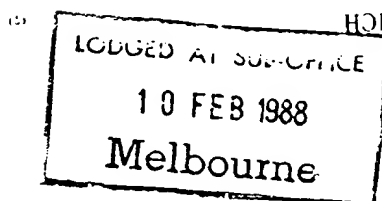
for a patent or similar protection made in (4) Federal Republic of Germany  
on 11th February 1987

My  
Our address for service is Messrs. Edwd. Waters & Sons, Patent Attorneys,

50 Queen Street, Melbourne, Victoria, Australia.

DATED this 9th day of February 19 88

(5) Signa-  
ture (s) of  
Applicant (s)  
or  
Seal of  
Company and  
Signatures of  
its Officers as  
prescribed by  
its Articles of  
Association



HOECHST AKTIENGESELLSCHAFT

by *[Signature]*  
D. B. Mischewski

registered Patent Attorney

COMMONWEALTH OF AUSTRALIAPatents Act 1952DECLARATION IN SUPPORT OF A CONVENTION APPLICATION UNDER PART XVI.  
FOR A PATENT.

In support of the Convention application made under Part XVI. of the Patents Act 1952 by HOECHST AKTIENGESELLSCHAFT of 45, Brüningstrasse, D-6230 Frankfurt/Main 80, Federal Republic of Germany for a patent for an invention entitled:

POLYPROPYLENE MOLDING MATERIAL

We, Johann-Heinrich Reuter, 4 Brüningstrasse, D-6230 Frankfurt/Main 80, Federal Republic of Germany, and Franz Laplow, 1 Sandweg, D-6500 Korbach, Federal Republic of Germany,

do solemnly and sincerely declare as follows:

1. We are authorized by HOECHST AKTIENGESELLSCHAFT the applicant for the patent to make this declaration on its behalf.

2. The basic application as defined by Section 141 of the Act was made in the Federal Republic of Germany under No. P 27 04 207.6 on February 11, 1967 by HOECHST AKTIENGESELLSCHAFT

3. a) Jürgen Helberg, 60 Johann-Strauß-Straße, D-6500 Korbach, Federal Republic of Germany  
 b) Gert Heufer, 131 Sperberstraße, D-6212 Bad Soden am Taunus  
 c) Friedrich Kloss, 112 Südring, D-6513 Mainz  
 d) Wolfgang Löw, 48 Lindensstraße, D-6597 Trebur  
 e) - d) Federal Republic of Germany

is/are the actual inventor(s) of the invention and the facts upon which HOECHST AKTIENGESELLSCHAFT

is entitled to make the application are as follows:

The said HOECHST AKTIENGESELLSCHAFT

is the assignee of the said

Jürgen Helberg, Gert Heufer, Friedrich Kloss, Wolfgang Löw

4. The basic application referred to in paragraph 2 of this Declaration was the first application made in a Convention country in respect of the invention the subject of the application.

DECLARED at Frankfurt/Main, Federal Republic of Germany

this 12th day of January 1988

To the Commissioner of Patents

HOECHST AKTIENGESELLSCHAFT

PAT 510

*[Signature]*  
Prokurist

ppa. Reuter

*[Signature]*  
Authorized signatory

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(12) PATENT ABSTRACT (11) Document No. AU-A-11485/88  
(19) AUSTRALIAN PATENT OFFICE  
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(54) Title

POLYPROPYLENE MOLDING MATERIAL.

(51)4 International Patent Classification

C08L 023/10 C08K 005/09 C08K 005/34 C08K 005/05  
C08K 003/34

(21) Application No. : 11485/88 (22) Application Date : 10.02.88

(30) Priority Data

(31) Number (32) Date (33) Country  
3704207 11.02.87 DE FEDERAL REPUBLIC OF GERMANY

(43) Publication Date : 18.8.88

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JURGEN HELBERG  
GERT HEUFER  
FRIEDRICH KLOOS  
WOLFGANG LOW

(74) Attorney or Agent

EDWD. WATERS & SONS

(57) Claim

1. A polypropylene molding material essentially consisting of a homopolymer of propylene or a copolymer of propylene with ethylene or butene, having a melt flow index MFI 230/5 of less than or equal to 5 g/10 min and containing 0.001 to 0.5% by weight, based on the molding material, of a nucleating agent from the group consisting of sodium benzoate, sodium montanate, calcium montanate, aluminum p-tert-butylbenzoate, quinacridone, naphthalimide, dibenzylidenesorbitol and ultrafine talc.
4. A process for improving the hardness and toughness of propylene moldings by adding nucleating agents to the molding material intended for the production of the moldings, wherein 0.001 to 0.5% by weight, based on the molding material, of a nucleating agent from the group consisting of sodium benzoate, sodium montanate, calcium montanate, aluminum p-tert-butylbenzoate, quinacridone, naphthalimide, dibenzylidenesorbitol and ultrafine talc is added to the molding material essentially consisting of a homopolymer of propylene or a

copolymer of propylene with ethylene or butene, having  
a melt flow index MFI 230/5 of less than or equal to  
5 g/10 min.

COMMONWEALTH OF AUSTRALIA

PATENTS ACT 1952/69

## COMPLETE SPECIFICATION

(ORIGINAL)

Class

Int. Class

Application Number:

Lodged:

Complete Specification Lodged:

Accepted:

Published:

Priority:

Related Art:

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Name of Applicant: HOECHST AKTIENGESELLSCHAFTAddress of Applicant: 45 Bruningstrasse, D-6230 Frankfurt/Main 80, Federal  
Republic of GermanyActual Inventor: JURGEN HELBERG, GERT HEUFER, FRIEDRICH KLOSS and  
WOLFGANG LOWAddress for Service: EDWD. WATERS & SONS,  
50 QUEEN STREET, MELBOURNE, AUSTRALIA, 3000.

Complete Specification for the invention entitled:

POLYPROPYLENE MOLDING MATERIAL

The following statement is a full description of this invention, including the best method of performing it known to the  
US



Description

Polypropylene molding material

The invention relates to a polypropylene molding material having improved processing characteristics and  
5 improved properties.

In the processing of polypropylene, the processing speed and the properties prove inadequate in many cases. There has therefore been no lack of attempts to overcome these deficiencies.

10 It is known that partially crystalline thermoplastics, such as polypropylene, can be advantageously affected in terms of processing characteristics and properties in many cases by adding suitable crystallization seeds, so-called nucleating agents. Nucleation leads to a finely  
15 spherulitic structure and a higher crystallization temperature.

The consequence of this is that the product to be prepared is dimensionally stable at a higher temperature and can therefore be removed from the mold at an earlier  
20 stage. This results in a substantial reduction in the cycle time during injection molding, in particular in the case of thick-walled parts, and in an increase in the take-off speed during extrusion.

The finely spherulitic structure leads to higher transparency and, because of the higher crystallization temperature, to increased crystallinity. At the same time there is an improvement in the hardness and rigidity of the moldings produced therefrom, but a reduction in the toughness properties. This undesired effect has considerably  
25 restricted the use of nucleating agents in many cases.  
30

It is known that by adding -quinacridone to a high molecular weight propylene homopolymer (cf. German Auslegeschrift 1,188,279) the hardness and toughness of the moldings produced therefrom is increased.  $\alpha$ - and  $\beta$ -quinacridones have only a slight effect; they increase only the hardness without producing any increase at all in the toughness.

Furthermore, the use of aluminum p-tert-butylbenzoate and dibenzylidenesorbitol as nucleating agents for propylene copolymer is known (cf. EP-A-137,482). However, talc is added to increase or maintain the low-temperature toughness. An improvement in the transparency and satisfactory rigidity and low-temperature toughness are achieved. Improvement of the hardness is not mentioned.

It was the object to find a propylene polymer for moldings having improved toughness and hardness.

It has been found that the object can be achieved by a propylene polymer which contains certain nucleating agents.

The invention therefore relates to a polypropylene molding material essentially consisting of a homopolymer or a copolymer of propylene with ethylene or butene, having a melt flow index MFI 230/5 of less than or equal to 5 g/10 min and containing 0.001 to 0.5% by weight, based on the molding material, of a nucleating agent from the group consisting of sodium benzoate, sodium montanate, calcium montanate, aluminum p-tert-butylbenzoate, quinacridone, naphthalimide, dibenzylidenesorbitol and ultrafine talc.

Suitable homopolymer and copolymers for the molding material according to the invention are homopolymers and copolymers of polypropylene with ethylene or butene which have a high molecular weight. Their melt flow index MFI 230/5 is less than or equal to 5 g/10 min (MFI

230/2.16  $\leq$  about 1 g/10 min), preferably less than or equal to 2 g/10 min (MFI 230/2.16  $\leq$  about 0.5 g/10 min).

The nucleating agents used are sodium benzoate, sodium montanate, calcium montanate, aluminum p-tert-butylbenzoate, quinacridone, naphthalimide, dibenzylidenesorbitol and ultrafine talc. Sodium benzoate, quinacridone and ultrafine talc are preferred, in particular sodium benzoate. The particle size of these nucleating agents is less than or equal to 20  $\mu\text{m}$ , preferably less than or equal to 10  $\mu\text{m}$ . The substances are added to the polymer in an amount of 0.001 to 0.5% by weight, preferably 0.001 to 0.15% by weight.

The molding material according to the invention can contain the conventional additives which facilitate processing and improve the physical and chemical properties. Examples of these are light stabilizers and heat stabilizers, antioxidants, antistatic agents, lubricants, anti-blocking agents and fillers and reinforcing agents, colored pigments, synthetic and natural resins, rubber-like products and flameproofing agents. The first group is present in the molding material in general in an amount of 0.01 to 5% by weight, based on the amount of polymer (+ filler). Fillers and reinforcing agents, colored pigments, synthetic and natural resins, rubber-like products and flameproofing agents are used in an amount corresponding to the particular requirements.

The nucleating agents are incorporated into the propylene polymer in a known manner, for example by means of a mixer, extruder, roll or kneader or by air homogenization, it being also possible to use a masterbatch of the nucleating agents for incorporation.

The molding material according to the invention not only results in improved toughness and improved hardness of the moldings in comparison with moldings obtained from

an identical molding material without the stated nucleating agents, but also possesses better processing characteristics. Thus, it permits shorter cycle times during injection molding and higher take-off speeds during extrusion of pipes and sections. Moreover, sticking of the plastic material to the mold is reduced during blow molding as a result of the formation of skin of higher crystallinity on the surface of the parison.

The molding material according to the invention can therefore particularly advantageously be used for producing thick-walled parts (for example cooling water reservoirs for motor vehicles) by injection molding, panels and pipes by extrusion and hollow articles, contoured articles (for example cases) and industrial articles (for example bumpers, spoilers, etc.) by blow molding.

If sodium benzoate is used as the nucleating agent, the presence of calcium stearate as an acid acceptor can be dispensed with. Calcium stearate furthermore has an adverse effect on nucleation with sodium benzoate.

The Examples which follow are intended to illustrate the invention:

Examples 1 to 7 and Comparative Examples A to O

Several high molecular weight ( $\text{MFI } 230/5 \leq 5 \text{ g/10 min}$ ) and low molecular weight ( $\text{MFI } 230/5 \geq 5 \text{ g/10 min}$ ) pulverulent homopolymers of propylene and copolymers of propylene with ethylene were mixed with various nucleating agents and, either as a powder or as granules, were processed to test specimens. In the same manner, test specimens which did not contain any nucleating agents were prepared for comparison. The hardness and the toughness of the test specimens were measured. The composition of the molding materials tested and the results of the measurements are summarized in Tables 1 and 2.

10 000 000

Table 1 (Comparative) Examples	Type	Starting mixtures			Nucleating agent	
		Initial PP powder APP (% by wt.)	C <sub>2</sub> H <sub>4</sub> (% by wt.)	MFI 230/5 (g/10 min)	Type	Concentration (% by wt.)
A	Homopolymer	4	-	0.26	-	-
1	"	"	-	"	Quinacridone	0.001
B	Copolymer	-	5	0.30	-	-
2	"	-	"	"	Quinacridone	0.001
C	"	-	6	0.25	-	-
3	"	-	"	"	Na benzoate	0.1
D	"	-	7	3.2	-	-
4	"	-	"	"	Al-p-tert-butyl- benzoate	0.05
E	"	-	13	4.5	-	-
5	"	-	"	"	Al-p-tert-butyl- benzoate	0.1
F	"	-	13	4.5	Ca montanate	-
6	"	-	"	"	Ultrafine talc	0.1
G	"	-	13	4.5	-	-
7	"	-	"	"	Al-p-tert-butyl- benzoate	0.1

Table 1 (continued)  
(Comparative)

Examples	Type	APP (% by wt.)	C <sub>2</sub> H <sub>4</sub> (% by wt.)	Initial PP powder		Nucleating agent	
				MF1 230/5	Type	Concentration	(% by wt.)
H1	Copolymer	-	9	18.5	-	-	-
H2	"	-	"	"	Na benzoate	0.3	0.3
I1	Homopolymer	2	-	55.4	-	-	-
I2	"	"	-	"	Ultrafine talc	0.3	0.3
K1	"	2	-	35.2	-	-	-
K2	"	"	-	"	Ultrafine talc	0.3	0.3
L1	Copolymer	-	9	21.1	-	-	-
L2	"	-	"	"	Ultrafine talc	0.3	0.3
M1	"	-	9	21	-	-	-
M2	"	-	"	"	Ultrafine talc	0.3	0.3

FINISHED PRODUCTS

Table 2

Comparative IZI Z30/5  
Examp.

	q/10	Form	Hardness/rigidity		Compressive strength		akv <sup>3)</sup> (N/mm <sup>2</sup> )		Strength in drop test		Drop test/sheets	
			BIH <sup>1)</sup>	FCM <sup>2)</sup>	1 min	bottles (N)	23°C	0°C	0°C	0°C	H 50 <sup>5)</sup>	-20°C
			(N/mm <sup>2</sup> )	(N/mm <sup>2</sup> )	value	3 mm	max.	max.	bottles <sup>4)</sup>	(cm)	(cm)	(cm)
A	0.94	Granules	65	-	-	-	-	16	-	-	-	-
1	0.93	"	68	-	-	-	-	25	-	-	-	-
B	0.83	Granules	51	-	-	-	-	12	-	-	-	-
2	0.85	"	53	-	-	-	-	21	-	-	-	-
C	0.25	Pulver	-	800	110	-	-	17	-	190	-	-
3	0.25	"	-	1150	165	-	-	30	-	250	-	-
II	0.6	Granules	-	850	123	-	-	15	-	150	-	-
3	0.6	"	-	1150	183	-	-	23	-	230	-	-
0	5.0	Granules	-	950	128	-	-	11	-	180	-	-
3	5.0	"	-	1160	210	-	-	11	-	200	-	-
D	6.0	Granules	56	-	161	193	-	10.7	-	186	-	-
4	6.2	"	62	-	179	213	-	14.1	-	167	-	-
E	3.7	Granules	54	877	-	-	-	34.8 18.1 14.4	-	-	240	130
5	3.8	"	58	1059	-	-	-	36.5 27.8 19.6	-	-	250	143



Table 2 (continued)

Comparative Examp.	MFI 230/5	Form	Hardness/rigidity													
			BIH <sup>1)</sup>	FOH <sup>2)</sup>	Compressive strength		akv <sup>3)</sup> (N/mm <sup>2</sup> )		Strength in drop test		Drop test/sheets					
					1 min value	3 mm max.	230°C	0°C	0°C	-20°C	0°C	-20°C				
													bottles (N)	bottles <sup>4)</sup>	bottles	H 50 <sup>5)</sup>
q/10	(N/mm <sup>2</sup> )	(N/mm <sup>2</sup> )	3 mm	max.	230°C	0°C	0°C	-20°C	0°C	-20°C	(cm)	(cm)				
F	3.7	Granules	54	877	-	-	34.8	18.1	14.4	-	240	130				
6	3.5	"	56	963	-	-	36.8	23.3	-	-	260	160				
G	3.7	Granules	54	877	-	-	34.8	18.1	14.4	-	240	130				
7	4.0	"	56	1060	-	-	37.3	24.6	-	-	-	130				
H1	21.8	Granules	58	1265	-	-	20.2	12.3	-	-	-	-				
H2	24.4	"	64	1409	-	-	15.8	10.8	-	-	-	-				
I1	62	Granules	80	1520	-	-	3.9	-	-	-	-	-				
I2	63	"	89	1710	-	-	3.1	-	-	-	-	-				
K1	165	Granules	85	1460	-	-	3.4	-	-	-	-	-				
L2	163	"	92	1730	-	-	2.9	-	-	-	-	-				
L1	59	Granules	62	1060	-	-	11.2	-	-	-	-	-				
L2	60	"	65	1200	-	-	9.0	-	-	-	-	-				
M1	160	Granules	57	980	-	-	7.4	-	-	-	-	-				
M2	157	"	62	1070	-	-	5.2	-	-	-	-	-				

Notes on Table 2

- 1) BIH: Ball indentation hardness, DIN 53,456, 4 mm sheet
  - 2) FCM: Flexural creep modulus, 1 min value, flexural creep test  $b = 5 \text{ N/mm}^2$
  - 3) akv: notched impact strength, corresponding to DIN 53,453, standard small bar with V notch
  - 4) Compressive strength/strength in drop test, bottles: (industrial test, Henkel)
- 10 a. Compressive strength
- A defined hollow article (parting sand container 700 ml, 35 g) is subjected to a load, and the force in N is measured at 3 mm and maximum deformation.
- 15 b. Progressive drop test
- Test temperature  $0^\circ\text{C}$
- 30 thermostated bottles are filled with water and placed on a drop table and allowed to fall onto a concrete floor, the height of fall being increased stepwise. The bottles which have remained intact are tested again with an increased height of fall, until the final bottle has been destroyed.
- 20 The height at which 50% of the bottles are destroyed (h 50 value) is determined.
- 5) Drop test, sheets ("Lukall", Hoechst)
- 25 Test temperatures  $23^\circ\text{C}$ ,  $0^\circ\text{C}$ ,  $-20^\circ\text{C}$ ,  $-40^\circ\text{C}$
- Falling ram 2 kg
- 6 sheets are tested in each case at different heights of fall. The end of the test corresponds to the height at which all 6 sheets are destroyed. The mean is calculated from this to give the mean height of fall.
- 30

~~Parent Cases:~~ THE CLAIMS DEFINE THE INVENTION AND ARE FORMULATED

1. A polypropylene molding material essentially consisting of a homopolymer of propylene or a copolymer of propylene with ethylene or butene, having a melt flow index MFI 230/5 of less than or equal to 5 g/10 min and containing 0.001 to 0.5% by weight, based on the molding material, of a nucleating agent from the group consisting of sodium benzoate, sodium montanate, calcium montanate, aluminum p-tert-butylbenzoate, quinacridone, naphthalimide, dibenzylidenesorbitol and ultrafine talc.
2. A molding material as claimed in claim 1, wherein the nucleating agent is sodium benzoate, aluminum p-tert-butylbenzoate, calcium montanate, quinacridone or ultrafine talc.
3. A molding material as claimed in claim 2, wherein the nucleating agent is sodium benzoate, quinacridone or ultrafine talc.
4. A process for improving the hardness and toughness of propylene moldings by adding nucleating agents to the molding material intended for the production of the moldings, wherein 0.001 to 0.5% by weight, based on the molding material, of a nucleating agent from the group consisting of sodium benzoate, sodium montanate, calcium montanate, aluminum p-tert-butylbenzoate, quinacridone, naphthalimide, dibenzylidenesorbitol and ultrafine talc is added to the molding material essentially consisting of a homopolymer of propylene or a copolymer of propylene with ethylene or butene, having a melt flow index MFI 230/5 of less than or equal to 5 g/10 min.
5. The process as claimed in claim 4, wherein the nucleating agent is sodium benzoate, aluminum p-tert-butylbenzoate, calcium montanate, quinacridone or ultrafine talc.

DATED this 9th day of February 1988.

BAECHST AKTIENGESellschaft

ELMD. WATERS & SONS,  
PATENT ATTORNEYS  
NEW YORK, N.Y. 10017

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